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Microstructure and Magnetic Property of L1o CoPt–20 at.% C Magnetic Thin Film

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ABSTRACT

The CoPt-20at.%C thin films of 20nm thickness were sputter-deposited in the form of $CoPt/C_n$ (n=1: carbon layer thickness=4nm; n=4: each carbon layer thickness=1nm) and were transformation-annealed at 650°C for various times. Carbon was found to dissolve into CoPt lattice and enlarge the c/a ratio of the ordered CoPt lattice. The amount of carbon dissolution increases with the decreasing carbon layer thickness at a given total carbon concentration.

The carbon dissolution larger than a critical amount can lead to a shift of the phase equilibrium of ordering and produce a stable fine two-phase mixture of ordered and disordered phases at the equi-atomic composition of Co:Pt. This results in a fine and uniform stagnant grain structure of about 20nm on annealing at 650°C. The carbon dissolution by increasing the c/a ratio of the ordered CoPt lattice reduces both the saturation magnetization and the magnetocrystalline anisotropy constant of the film and leads to a reduction of coercivity of CoPt films.

INTRODUCTION

The ordered L1o CoPt granular film is a potential candidate for ultra high density recording media because of its high magneto-crystalline anisotropy constant. This film is however known to be prone to the occurrence of excessive grain growth during transformation annealing and to high transition noise because of lack of magnetic granular isolation. To solve this problem, the composite film of L1o granular CoPt particles imbedded in the carbon matrix has been the subject of extensive studies[1,2]. The composite film was fabricated either by co-sputtering Co, Pt, and C[1] or by sputtering in the form of multilayers (CoPt/C)n [2] followed by annealing.

We were interested in exploring the possibility of fabricating optimum monolithic, instead of composite, CoPt(C) granular film by incorporating a minor amount of carbon in the form of $CoPt/C_n$(n=1, 4). We investigated the effect of C mixing on the grain structure and magnetic properties of L1o CoPt granular film. Particular attention was paid to the mixing behavior of

carbon layer with CoPt layer by dividing the given carbon layer into several thin layers for mixing experiments.
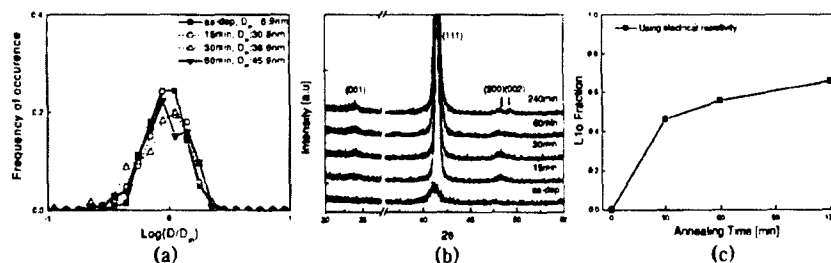
EXPERIMENTS

Equiatomic CoPt film was RF-sputter deposited on the thermally oxidized Si substrate at room temperature under the Ar pressure 5 mtorr in a vacuum chamber with a base pressure less than 1×10^{-6} torr. The intercalated carbon layer was DC-sputter deposited using carbon target. The total film thickness of CoPt layer was fixed to 20 nm and that of carbon layer was fixed to 4 nm, which amounts to about 20 at.% C. To investigate the effect of thickness effect on the carbon mixing with CoPt lattice, alternate sputtering was carried out to fabricate a mutilayer structure of CoPt/C_n (n=1, 4) by dividing the total carbon layer (n=1: thickness=4nm) into 4 thin carbon layers (n=4: thickness=1nm). The films were annealed at 650°C for various times in a vacuum furnace. The ordering transformation was studied using X-ray diffractometer and TEM DF technique. The grain structure was characterized using TEM and the in-plane grain size distribution was measured using an image analyzer. The number of counted grains was 200-500. The magnetic hysteresis loop was measured using VSM.

RESULTS AND DISCUSSION

The examination of in-plane grain structure CoPt thin film (n=0) showed that the grain grows rapidly during the initial period of annealing at 650°C and tends to be stagnant at a later stage. The addition of carbon of 20 at.% into CoPt film in the form of multilayer CoPt/C_n (n=1, 4) was observed to greatly retard the initial growth rate. Furthermore, the stagnant grain structure was found to be greatly dependent on the number of divided carbon layers, n at a given total carbon thickness, namely on the thickness of each carbon layer.

The grain size distribution (GSD) of 20nm CoPt thin films is plotted in terms of frequency vs. logarithm of the normalized grain size (Fig.1a). The as–deposited film exhibits a lognormal grain size distribution. With the progress of annealing, the grain population near the peak diminishes with a concurrent rise of grain population at the leading edge, i.e., the side of small grains. This eventually leads to a bimodal-like grain size distribution on annealing for 30 min. Further progress of annealing recovers a monomodal grain size distribution and the GSD approaches a lognormal distribution. This variation is qualitatively in correspondence with that of the transformation fraction of L1o ordered CoPt phase with annealing. The X-ray diffraction study (Fig.1b) indicated that the transformation rate is rapid at 30 min of annealing and the rate
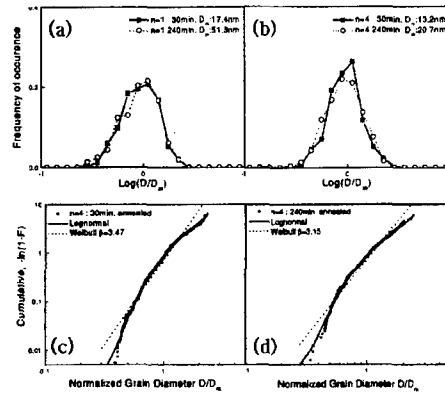
Figure 1. Plot of the frequency of occurrence vs. logarithm of the normalized grain size with respect to the average grain size D_m (a) ; XRD patterns of of 20nm CoPt (n=0) films as-deposited and annealed at 650°C for various times (b) ; Plot of L1o fraction vs. annealing times at 650°C in 40nm CoPt (n=0) films.

slows down after 60 min of annealing. This result was further confirmed in the measurement of transformed volume fraction of ordered phase for 40nm CoPt thin films using electrical resistivity technique (Fig.1c).

This result shows that the ordering transformation occurs by nucleation of ordered clusters and their growth. It appears that these clusters do not grow fast because of the presence of interface between ordered and disordered phases and that small particles were found at 30 min of annealing. The presence of small ordered particles was in fact observed in a TEM DF study in agreement with the previous work by Coffey et al. [3].

Figure 2 shows the GSD in 20nm CoPt/C$_n$ (n= 1, 4) thin films. The GSD rather resembles to a mono-modal than a bi-modal distribution, which is particularly true for thin film with n=4 (each carbon layer thickness=1nm): there appears to be some faint trace of bimodal distribution for film with n=1 (carbon layer thickness=4nm) for a long time annealing. The cumulative-frequency plot showed that the GSD's are all close to a lognormal distribution except at the region of small grains, i.e.: the leading edge. The leading edge with a long time annealing always better fits to the lognormal distribution. It is to remark here that the GSD of the film with n = 4 approaches a nearly perfect lognormal distribution on annealing for 240min, although the average grain size at this condition is only about the size of film thickness, 20nm. This is a premature stagnation of grain structure in view of the well established fact that the grain stagnation usually occurs at D ~ 2.5 t [4].

We believe that this premature stagnation of grain structure arises from the formation of a stable two-phase mixture of ordered and disordered phases in CoPt/C$_4$ films, which is also

Figure 2. Plot of the frequency of occurrence vs. logarithm of the normalized grain size of the CoPt/C_n films with n=1 (a) and n=4 (b) and the cumulative-frequency plot of the CoPt/C_4 films for different annealing times (c, d).

responsible for the formation of very fine and uniform GSD's (Fig.2). The reason for the exceptional stability in the n=4 film is that the ordering transformation is extremely limited over a long period of annealing time in the film with n=4 (carbon one-layer thickness = 1nm) unlike from the film with n=1 (carbon one-layer thickness = 4nm) despite the fact that the total carbon thickness is the same in both films. The X- ray diffraction study (Fig.3a) clearly showed the difference: the ordering transformation is greatly suppressed in the film of CoPt/C_4 as compared with the film of CoPt/C_1 although the total carbon thickness is the same. In addition, the (111) peak is shown to significantly shift to a larger angle in the n=1 film as compared with the CoPt film, indicating that carbon dissolves into CoPt lattice. The result further shows that the (111) peak in the n=4 film experiences a peak separation due to ordering transformation, a small secondary ordered peak appearing at the high angle side of principal peak: this indicates that the transformation is incomplete in this film even after a sufficient annealing. Furthermore, the angular position of this secondary ordered peak is clearly at a larger angle than that of (111) peak in the film with n=1. This clearly indicates that the amount of dissolved carbon increases with increasing n, that is, with the decreasing carbon layer thickness. The (001) ordered peak, however, appears not to change with carbon addition, although its accurate position is somewhat obscure because of a low intensity.

From the information of the X-ray diffraction studies, we can conclude that the carbon dissolves into ordered L1o CoPt lattice and that the carbon solubility increases with decreasing
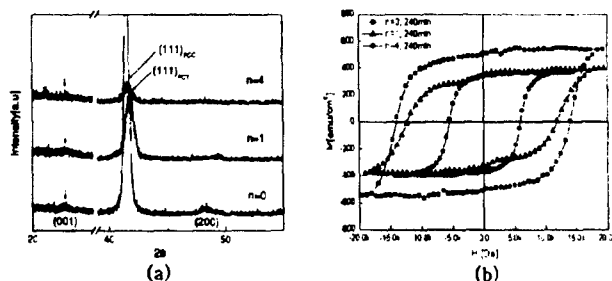
Figure 3. XRD patterns of CoPt/C$_n$ (n=0, 1, 4) films annealed for 30 min at 650°C (a) and in-plane magnetic hysteresis loops of CoPt/C$_n$ (n=0, 1, 4) films annealed for 240 min at 650°C (b).

individual carbon layer thickness at a given total carbon thickness. The dissolved carbon makes the c/a ratio bigger by reducing a and the c/a increases with the number of divided carbon layer, i.e., with the diminishing individual carbon layer thickness at a given total thickness. The approximate calculation of c/a ratio from the present X-ray data indicated that the c/a ratios are 0.945 for n=0, 0.957 for n=1 and 0.964 for n=4.

We believe that a large increase of c/a ratio in the n=4 film is primarily responsible for a great suppression of ordering transformation and for the formation of stable two-phase mixture of ordered and disordered phases. The origin of the formation of a stable two-phase mixture is believed not to be the kinetic one but rather to be thermodynamic one, in view of its great thermal stability. This result suggests that Tc shifts toward a higher temperature and Pt-rich composition so that the film with n=4 is found within the two-phase region at 650°C. It is to remark here that this does not happen in the film with n=1 although the overall carbon composition is the same. This suggests that a critical amount of carbon dissolution is necessary in order to significantly shift the equilibrium phase field of Co-Pt system.

The fact that carbon dissolves into L1o CoPt lattice and that the amount of carbon dissolution depends on the carbon layer thickness is also well manifested in the magnetic hysteresis loops of CoPt films. The measurement of hysteresis loops (Fig.3b) showed that the coercivity, among others, is reduced as a result of carbon incorporation into the ordered CoPt lattice and the amount of reduction strongly depends on the thickness of initial carbon layer. We believe that this is because the increase of c/a ratio of ordered phase with carbon incorporation reduces the degree of ordering of L1o CoPt lattice as well as its transformation fraction. Both factors contribute to a reduction of effective Ku of CoPt film and lead to a reduction of its

coercivity. The carbon dissolution into the ordered CoPt lattice also reduces the Ms of L1o CoPt phase. A similar Ms value is observed in the films with n=1 and n=4. This is because the film with n=4 consists of ordered and disordered phases, the latter phase exhibiting a larger Ms value. All of these results suggest that the CoPt –20at.%C film is one of promising candidates for solving the excessive grain growth and transition noise problems of the high density magnetic recording media based on CoPt alloys.

CONCLUSIONS

The CoPt-20at.%C thin films of 20nm thickness were sputter-deposited in the form of CoPt/C_n (n=1: carbon layer thickness=4nm; n=4: each carbon layer thickness=1nm) and were transformation-annealed at 650°C for various times. Carbon was found to dissolve into CoPt lattice and enlarge the c/a ratio of the ordered CoPt lattice. The amount of carbon dissolution increases with the decreasing carbon layer thickness at a given total carbon concentration.
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ACKNOWLEDGMENTS

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